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THEORY OF SECOND HARMONIC GENERATION IN PRESENCE OF DIFFRACTION, BEAM WALK-OFF AND PUMP DEPLETION

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Theory of Second Harmonic Generation in presence of Diffraction, Beam Walk-Off and Pump Depletion

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ABSTRACT

Integral expressions for the pump and generated fields are presented here for the case of second harmonic generation of a focused Gaussian pump beam incident on a nonlinear crystal . The birefringent walk-off of the generated beam and the effect of pump depletion are included in the theory.

1. INTRODUCTION

The theory of second harmonic generation (SHG) of a focused Gaussian beam in the presence of beam walk-off presented by Boyd and Kleinman[1] has been widely used in the nonlinear optics community. A limitation of their method is the exclusion of pump depletion which makes it inapplicable to the case of high conversion efficiency. The numerical method developed by Smith[2] and recent work by Wang and Weiner[3] and by Kasamatsu, Kubomura and Kan[4] extend the theory of Boyd and Kleinman to include the effect of pump depletion. We present here an alternate technique to calculate the spatial distribution of the pump and the SHG fields in presence of diffraction, linear absorption, phase mismatch, beam walk-off and pump depletion. The field distributions are expressed as multiple integrals, which are considerably simplified for the case of collimated beams, i.e., when the spread of the pump beam due to diffraction inside the crystal can be ignored. This is often the case when high peak power pulsed lasers are used as pump beams because the pump is intentionally kept collimated through the nonlinear optical medium to avoid damage to the material.

In the theory presented here the incident pump beam is assumed to be focused at the crystal center - this restriction can be easily removed later. The aim of this work is to describe the general approach taken and to provide expressions for the electric fields expanded up to fifth power in the second-order nonlinear optical coefficient, d. The method described here is an extension of an early work on optical parametric oscillation (OPO)[5] and can be readily applied to other nonlinear optical processes of current interest, such as pump resonant SRO or resonant second harmonic generation (SHG). As pointed out in ref.3, the Green's function method is easily extended to the time dependent case as well. However, a limitation of the perturbative approach used here is that multiple integrals need to be evaluated, which becomes computationally time consuming when six, eight or higher dimensional integrals are involved.

In the following sections, the derivation of the relevant wave equations is provided first, to set up the starting equations. Then, the Green's function method of solution in terms of a power series in the nonlinear coefficient is described, and the expressions for the pump and the SHG beam electric fields are evaluated for a few terms.

2. THE STARTING EQUATIONS:

The notations for electric fields and nonlinear polarizations used here will generally follow the convention adopted in the *Handbook of Nonlinear Optics*[6] (using the SI system of units). The subscripts p and s denote the pump and the SHG fields, respectively. The fields are assumed to be linearly polarized, and only the scalar components of the electric fields are considered in the nonlinear interaction. The equation 16.4.5 in *Quantum Electronics*[7] is taken as the starting point of the analysis here:

$$\nabla^2 \mathbf{E} = \mu_0 \sigma \frac{\partial \mathbf{E}}{\partial t} + \mu_0 \epsilon \frac{\partial^2 \mathbf{E}}{\partial t^2} + \mu_0 \frac{\partial^2}{\partial t^2} \mathbf{P_{NL}} \tag{1}$$

1

The electric fields at the pump and the SHG frequencies (ω_p and ω_s, respectively) are written as

$$E^{\omega_p}(\mathbf{r}, t) = E_p(\mathbf{r})e^{-i\omega_p t} + cc \tag{2}$$

$$E^{\omega_s}(\mathbf{r}, t) = E_s(\mathbf{r})e^{-i\omega_s t} + cc \tag{3}$$

and the total nonlinear polarization as

$$P_{NL}(\mathbf{r}, t) = (\mathcal{P}_{NL}^{\omega_p}(\mathbf{r})e^{-i\omega_p t} + cc) + (\mathcal{P}_{NL}^{\omega_s}(\mathbf{r})e^{-i\omega_s t} + cc) \tag{4}$$

Following ref. 6 (chapter 2) we assume that the nonlinear polarization terms at the pump and the SHG frequencies can be written as

$$\mathcal{P}_{NL}^{\omega_p}(\mathbf{r}) = 4d\epsilon_0 E_s(\mathbf{r})E_p^*(\mathbf{r}) \tag{5}$$

$$\mathcal{P}_{NL}^{\omega_s}(\mathbf{r}) = 2d\epsilon_0 E_p(\mathbf{r})^2 \tag{6}$$

Inserting equations 2,3,4,5 and 6 in equation 1, and collecting terms oscillating at frequencies ω_p and ω_s, we obtain,

$$(\nabla^2 + k_{p_1}^2)E_p(\mathbf{r}) = -\mu_0\omega_p^2\mathcal{P}_{NL}^{\omega_p}(\mathbf{r}) \tag{7}$$

$$(\nabla^2 + k_{s_1}^2)E_s(\mathbf{r}) = -\mu_0\omega_s^2\mathcal{P}_{NL}^{\omega_s}(\mathbf{r}) \tag{8}$$

where

$$k_{p_1}^2 = k_p^2 + i\omega_p\mu_0\sigma_p \qquad k_{s_1}^2 = k_s^2 + i\omega_s\mu_0\sigma_s \tag{9}$$

In the above, it has been assumed that in equation 1, $\epsilon_{p,s} = \epsilon_0 n_{p,s}^2$ and that the wavevectors are $k_{p,s} = n_{p,s}\omega_{p,s}/c$ where n_p and n_s denote the refractive indices of the medium at the two frequencies.

3. GREEN'S FUNCTION SOLUTION

The solution to the inhomogeneous differential equations 7 and 8 above is obtained using the Green's function method:

$$E_p(\mathbf{r}) = -\frac{\mu_0\omega_p^2}{4\pi}\int d\mathbf{r}'\frac{e^{ik_{p_1}R_p}}{R_p}\mathcal{P}_{NL}^{\omega_p}(\mathbf{r}) \tag{10}$$

$$E_s(\mathbf{r}) = -\frac{\mu_0\omega_s^2}{4\pi}\int d\mathbf{r}'\frac{e^{ik_{s_1}R_s}}{R_s}\mathcal{P}_{NL}^{\omega_s}(\mathbf{r}) \tag{11}$$

where R_p and R_s denote the magnitudes of the displacement vectors connecting the points $\mathbf{r}'$ and $\mathbf{r}$ for the pump and the SHG fields, respectively. The pump beam is assumed to propagate in the medium as an ordinary wave, so that

$$\begin{aligned} R_p &= |\mathbf{r} - \mathbf{r}'| \\ &= \sqrt{(x-x')^2 + (y-y')^2 + (z-z')^2} \end{aligned} \tag{12}$$

If the SHG field propagates through the nonlinear crystal as an extraordinary wave, say with a walk-off angle denoted by ρ, and if the Cartesian coordinates are chosen such that the propagation vector of the SHG field lies in the x, z plane and is at angle ρ with the z axis, we have [1]

$$R_s = \sqrt{\{(x - \rho z) - (x' - \rho z')\}^2 + (y - y')^2 + (z - z')^2} \tag{13}$$

For small absorption, equation 11 can be re-written as:

$$k_{p_1} \cong k_p + i\frac{\alpha_p}{2} \qquad k_{s_1} \cong k_s + i\frac{\alpha_s}{2} \tag{14}$$

where $\alpha_{p,s} = \sigma\sqrt{\mu_0/\epsilon_{p,s}}$ denote the the linear absorption coefficients. The Fresnel approximation

$$R_p \cong (z - z') + \frac{1}{2(z - z')}\{(x - x')^2 + (y - y')^2\} \tag{15}$$

2

and

$$R_s \cong (z - z') + \frac{1}{2(z - z')}\{(x_a - x')^2 + (y - y')^2\} \tag{16}$$

is used in the exponents, with $x_a = x - \rho(z - z')$ and the approximation $R_p, R_s \cong (z - z')$ is used in the denominators. Equations 10 and 11 then reduce to

$$E_p(\mathbf{r}) = -\frac{\mu_0 \omega_p^2}{4\pi} \int \frac{dx' dy' dz'}{z - z'}$$
$$e^{ik_p(z-z') - \frac{\alpha_p}{2}(z-z') + \frac{ik_p}{2(z-z')}\{(x-x')^2 + (y-y')^2\}} \mathcal{P}_{NL}^{\omega_p}(\mathbf{r}') \tag{17}$$

and

$$E_s(\mathbf{r}) = -\frac{\mu_0 \omega_i^2}{4\pi} \int \frac{dx' dy' dz'}{z - z'}$$
$$e^{ik_s(z-z') - \frac{\alpha_s}{2}(z-z') + \frac{ik_s}{2(z-z')}\{(x_a-x')^2 + (y-y')^2\}} \mathcal{P}_{NL}^{\omega_s}(\mathbf{r}') \tag{18}$$

The incident pump beam is assumed to have Gaussian cross-sections and to be focused at the center of a nonlinear medium of length l. Assuming the origin of the coordinate system to be at the incident face of the medium, the amplitude $E_p^{(0)}(\mathbf{r})$ of the incident pump field can be written as

$$E_p^{(0)}(\mathbf{r}) = \frac{E_{p0}}{1 + 2i(z - \frac{l}{2})/b_p} e^{ik_p z - \alpha_p z/2} e^{-(x^2 + y^2)/\{w_{0p}^2(1 + 2i(z - \frac{l}{2})/b_p\}} \tag{19}$$

where $b_p \equiv k_p w_{0p}^2$ denotes the confocal parameter. In the first order approximation, i.e., when pump depletion is ignored, it is assumed that the pump field E_p remains equal to $E_p^{(0)}$, a nonlinear polarization at frequency ω_s is generated through equation 6, which gives rises to an electric field $(dE_s^{(1)})$ at frequency ω_s through equation 11. This generated SHG field interacts with the pump field E_p^0 to generate a nonlinear polarization through equation 5, which generates (through equation 10) a component of the electric field at the pump frequency (denoted by, say, $d^2 E_p^{(2)}$). Following this procedure, a series of SHG and pump field terms, respectively odd and even powered in d, can be obtained. That is, in general, the SHG and the pump fields can be written as

$$E_s = dE_s^{(1)} + d^3 E_s^{(3)} + d^5 E_s^{(5)} \ldots \tag{20}$$

$$E_p = E_p^{(0)} + d^2 E_p^{(2)} + d^4 E_p^{(4)} + d^6 E_p^{(6)} \ldots \tag{21}$$

Substituting these series in the equations 5 and 6, the nonlinear polarization terms can also be expanded in power series in d:

$$\mathcal{P}_{NL}^{\omega_s} = \mathcal{P}_{NL}^{s(1)} + \mathcal{P}_{NL}^{s(3)} + \mathcal{P}_{NL}^{s(5)} \ldots \tag{22}$$

$$\mathcal{P}_{NL}^{\omega_p} = \mathcal{P}_{NL}^{p(2)} + \mathcal{P}_{NL}^{p(4)} + \mathcal{P}_{NL}^{p(6)} \ldots \tag{23}$$

where

$$\mathcal{P}_{NL}^{s(1)} = 2d\epsilon_0 E_p^{(0)2} \tag{24}$$

$$\mathcal{P}_{NL}^{s(3)} = 4d^3 \epsilon_0 E_p^{(0)} E_p^{(2)} \tag{25}$$

$$\mathcal{P}_{NL}^{s(5)} = 2d^5 \epsilon_0 (E_p^{(2)2} + 2E_p^{(4)} E_p^{(0)}) \tag{26}$$

and

$$\mathcal{P}_{NL}^{p(2)} = 4d^2 \epsilon_0 E_s^{(1)} E_p^{(0)*} \tag{27}$$

$$\mathcal{P}_{NL}^{p(4)} = 4d^4 \epsilon_0 (E_s^{(1)} E_p^{(2)*} + E_s^{(3)} E_p^{(0)*}) \tag{28}$$

$$\mathcal{P}_{NL}^{p(6)} = 4d^6 \epsilon_0 (E_s^{(5)} E_p^{(0)*} + E_s^{(3)} E_p^{(2)*} + E_s^{(1)} E_p^{(4)*}) \tag{29}$$

Using equations 19 and 24, and substituting in 18, the first term on the right hand side of eqn. 20 can be determined. Using this expression for $dE_i^{(1)}$ and eqn. 19 in eqn. 27, the $d^2 E_p^{(2)}$ term in eqn. 21 can be evaluated from eqn. 17. With this $d^2 E_p^{(2)}$ and the E_s from eqn. 19 substituted in eqn. 25, equation 17 can be used for the evaluation of the $d^3 E_s^{(3)}$ term in Eqn. 20, and so on. Before embarking on the explicit determination of each of these terms, in the next section several parameters are defined, and a new notation is introduced which allows all the spatial integrations to be performed in dimensionless units.

3.1. Notation

The parameter $\xi_p \equiv l/b_p$ where l denotes the length of the nonlinear medium is used to characterize the amount of focusing of the pump beam. The phase mismatch between the nonlinear polarization beam and the generated SHG wave is denoted by

$$\Delta k \equiv 2k_p - k_s. \tag{30}$$

Similarly, the mismatch in linear absorption between the nonlinear polarization and the SHG beam is proportional to

$$\Delta \alpha \equiv \alpha_p - \alpha_s/2. \tag{31}$$

In addition, the following parameters are defined:

$$p \equiv x/w_{op} \quad q \equiv y/w_{op} \quad t \equiv z/l \tag{32}$$

$$p_1 \equiv x'/w_{op} \quad q_1 \equiv y'/w_{op} \quad t_1 \equiv z'/l \tag{33}$$

$$p_a \equiv x_a/w_{op} \quad W_1 \equiv \rho l/w_{0p} \tag{34}$$

where W_1 is a dimensionless parameter which provides a measure of the amount of walk-off of the SHG wave as it propagates through the crystal. Also, defining the variables a_p and $d_p(t)$ as

$$a_p \equiv 1 - i\xi_p$$
$$d_1(t) \equiv a_p + 2i\xi_p t \tag{35}$$

the incident pump field can be expressed as:

$$E_p^{(0)}(\mathbf{r}) = \frac{E_{p0}}{d_1(t)} e^{ik_p lt - \alpha_p lt/2} e^{-(p^2+q^2)/d_1(t)} \tag{36}$$

$$\tag{37}$$

4. THE EXPRESSIONS FOR THE ELECTRIC FIELD TERMS

4.1. The first order SHG field term

Using Eqns. 18,19 and 24, the first order signal field term $dE_s^{(1)}$ is obtained (after integrating over the transverse coordinates):

$$dE_s^{(1)}(\mathbf{r}) = \kappa E_{p0} f_s^{(1)}(\mathbf{r}) \tag{38}$$

where

$$\kappa \equiv -\frac{2i\pi dl E_{p0}}{\lambda_s n_s} \tag{39}$$

and

$$f_s^{(1)}(\mathbf{r}) \equiv e^{(ik_s - \alpha_s/2)lt} \int_0^t dt_1 \frac{e^{(i\Delta k - \Delta\alpha)lt_1} e^{-(p_a{}^2+q^2)\frac{2}{d_2(t,t_1)}}}{d_1(t_1) d_2(t,t_1)}. \tag{40}$$

Here $p_a = p - W_1(t - t_1)$. The function $d_2(t, t_1)$ is defined as

$$d_2(t, t_1) \equiv d_1(t_1) + 2ik\xi_p(t - t_1), \tag{41}$$

where $k \equiv 2k_p/k_s$.

4

4.2. The second order pump field term

Using Eqns 38,19, 27 and 17, the second order pump field term $d^2 E_p^{(2)}$ can be obtained (after integrating over the transverse coordinates):

$$d^2 E_p^{(2)}(\mathbf{r}) = -\frac{n_s}{n_p} \mid \kappa \mid^2 f_p^{(2)}(\mathbf{r}) \tag{42}$$

where

$$
\begin{aligned}
f_p^{(2)}(\mathbf{r}) \;\equiv\;& e^{(ik_p - \alpha_p/2)lt} \int_0^t dt_1 e^{(-i\Delta k - \alpha_s/2)lt_1} \\
&\times \int_0^{t_1} dt_2 e^{(-i\Delta k - \Delta\alpha)lt_2} \\
&\times \frac{e^{-(p^2+q^2)a_2 + 2pb_2 + c_2}}{d_1(t_4)d_8(t,t_1,t_2,t_3,t_4)}
\end{aligned}
\tag{43}
$$

with

$$a_2(t, t_1, t_2) \equiv \frac{d_3(t_1, t_2)}{d_4(t, t_1, t_2)}, \tag{44}$$

$$b_2(t, t_1, t_2) \equiv 2W_1(t_1 - t_2)\frac{d_1(t_1)^*}{d_4(t, t_1, t_2)}, \tag{45}$$

and

$$c_2(t, t_1, t_2) \equiv -2W_1{}^2(t_1 - t_2)^2 \left(\frac{d_{2a}(t, t_1)}{d_4(t, t_1, t_2)}\right), \tag{46}$$

Other functions used above are defined as

$$d_{2a}(t, t_1) \equiv d_1(t_1)^* + 2i\xi_p(t - t_1) \tag{47}$$

$$d_3(t_1, t_2) \equiv d_2(t_1, t_2) + 2d_1(t_1)^* \tag{48}$$

$$d_4(t, t_1, t_2) \equiv d_1(t_1)^* d_2(t_1, t_2) + 2i\xi_p(t - t_1)d_3(t_1, t_2) \tag{49}$$

4.3. The third order SHG field term

Using Eqns. 19, 42, 25 and 18, the third order SHG field term $d^3 E_s^{(3)}$ can be obtained (after integrating over the transverse coordinates):

$$d^3 E_s^{(3)}(\mathbf{r}) = -\left(\frac{n_s}{n_p}\right)^2 \kappa \mid \kappa \mid^2 f_s^{(3)}(\mathbf{r}) \tag{50}$$

where

$$
\begin{aligned}
f_s^{(3)}(\mathbf{r}) \;\equiv\;& e^{(ik_s - \alpha_s/2)lt} \int_0^t dt_1 e^{(i\Delta k - \Delta\alpha)lt_1} \\
&\times \int_0^{t_1} dt_2 e^{(-i\Delta k - \alpha_s/2)lt_2} \int_0^{t_2} dt_3 e^{(i\Delta k - \Delta\alpha)lt_3} \\
&\times \frac{e^{-(p_a{}^2+q^2)a_3 + 2p_a b_3 + c_3}}{d_1(t_3)d_6(t, t_1, t_2)}
\end{aligned}
\tag{51}
$$

where

$$a_3(t, t_1, t_2) \equiv \frac{d_5(t_1, t_2, t_3)}{d_6(t, t_1, t_2, t_3)}, \tag{52}$$

$$b_3(t, t_1, t_2) \equiv 2W_1(t_2 - t_3)\frac{d_1(t_2)^* d_1(t_1)}{d_6(t, t_1, t_2, t_3)}, \tag{53}$$

5

and

$$c_3(t, t_1, t_2) \equiv W_1{}^2 (t_2 - t_3)^2 \frac{g_1(t, t_1, t_2)}{d_4(t, t_1, t_2) d_6(t, t_1, t_2, t_3)},$$ (54)

with

$$d_5(t_1, t_2, t_3) \equiv d_4(t_1, t_2, t_3) + d_1(t_1) d_3(t_2, t_3)$$ (55)

$$d_6(t, t_1, t_2, t_3) \equiv d_1(t_1) d_4(t_1, t_2, t_3) + ik\xi_p(t - t_1) d_5(t_1, t_2)$$ (56)

and

$$g_1(t, t_1, t_2, t_3) = 4ik\xi_p(t - t_1) d_1(t_1) d_1(t_2)^{*2} - 2d_{2a}(t_1, t_2) d_6(t, t_1, t_2, t_3)$$ (57)

4.4. The fourth order pump field terms

Using Eqns. 38, 42, 50 and 19 in 28 and 17, the fourth order pump field term $d^4 E_p^{(4)}$ is obtained as a sum of two terms $d^4 E_{p1}^{(4)}$ and $d^4 E_{p2}^{(4)}$:

$$d^4 E_{p1}^{(4)}(\mathbf{r}) = 2(\frac{n_s}{n_p})^2 \mid \kappa \mid^4 E_{p0} f_{p1}^{(4)}(\mathbf{r})$$ (58)

$$d^4 E_{p2}^{(4)}(\mathbf{r}) = (\frac{n_s}{n_p})^2 \mid \kappa \mid^4 E_{p0} f_{p2}^{(4)}(\mathbf{r})$$ (59)

The expressions for $f_{p1}^{(4)}$ and $f_{p2}^{(4)}$ are given in the next two subsections.

4.4.1. $f_{p1}^{(4)}$

$$
\begin{aligned}
f_{p1}^{(4)}(\mathbf{r}) &\equiv e^{(ik_p - \alpha_p/2)lt} \int_0^t dt_1 e^{(-i\Delta k - \alpha_s/2)lt_1} \int_0^{t_1} dt_2 e^{(i\Delta k - \Delta\alpha)lt_2} \\
&\times \int_0^{t_2} dt_3 e^{(-i\Delta k - \alpha_s/2)lt_3} \int_0^{t_3} dt_4 e^{(i\Delta k - \Delta\alpha)lt_4} \\
&\times \frac{e^{-(p^2 + q^2)a_{41} + 2pb_{41} + c_{41}}}{d_1(t_4) d_8(t, t_1, t_2, t_3, t_4)}
\end{aligned}
$$ (60)

where

$$a_{41}(t, t_1, t_2, t_3, t_4) \equiv \frac{d_7(t_1, t_2, t_3, t_4)}{d_8(t_1, t_2, t_3, t_4)},$$ (61)

$$b_{41}(t, t_1, t_2, t_3, t_4) \equiv W_1 \frac{d_1(t_1)^* g_2(t_1, t_2, t_3, t_4)}{d_8(t, t_1, t_2, t_3, t_4)},$$ (62)

and

$$c_{41}(t, t_1, t_2, t_3, t_4) \equiv W_1{}^2 \{g_3(t_1, t_2, t_3, t_4) + g_4(t, t_1, t_2, t_3, t_4)\}$$ (63)

with

$$g_2(t_1, t_2, t_3, t_4) \equiv (t_1 - t_2) d_5(t_2, t_3, t_4) - 2(t_3 - t_4) d_1(t_3)^* d_1(t_2)$$ (64)

$$g_3(t_1, t_2, t_3, t_4) \equiv \frac{g_{3n}(t_1, t_2, t_3, t_4)}{g_{3d}(t_1, t_2, t_3, t_4)}$$ (65)

$$g_4(t, t_1, t_2, t_3, t_4) \equiv \frac{g_{4n}(t, t_1, t_2, t_3, t_4)}{g_{4d}(t, t_1, t_2, t_3, t_4)}.$$ (66)

$$
\begin{aligned}
g_{3n}(t_1, t_2, t_3, t_4) &= (t_3 - t_4)^2 g_1(t_1, t_2, t_3, t_4) \\
&- (t_1 - t_2)^2 d_4(t_2, t_3, t_4) d_5(t_2, t_3, t_4)
\end{aligned}
$$ (67)

$$g_{3d}(t_1, t_2, t_3, t_4) = d_4(t_2, t_3, t_4) d_6(t_1, t_2, t_3, t_4)$$ (68)

$$g_{4n}(t, t_1, t_2, t_3, t_4) = 2i\xi_p(t - t_1) d_1(t_1)^* g_2(t_1, t_2, t_2, t_3)^2$$ (69)

6

$$g_{4d}(t_1, t_2, t_3, t_4) = d_6(t_1, t_2, t_3, t_4)d_8(t, t_1, t_2, t_3, t_4). \tag{70}$$

The functions d_7 and d_8 are given by

$$d_7(t_1, t_2, t_3, t_4) \equiv d_6(t_1, t_2, t_3, t_4) + d_1(t_1)^* d_5(t_2, t_3, t_4) \tag{71}$$

and

$$d_8(t_1, t_2, t_3, t_4) \equiv d_1(t_1)^* d_6(t_1, t_2, t_3, t_4) + 2i\xi_p(t - t_1)d_7(t_1, t_2, t_3, t_4) \tag{72}$$

4.4.2. $f_{p2}^{(4)}$

$$
\begin{aligned}
f_{p2}^{(4)}(\mathbf{r}) \equiv\ & e^{(ik_p - \alpha_p/2)lt} \int_0^t dt_1 e^{(-i\Delta k - \alpha_s/2)lt_1} \int_0^{t_1} dt_2 e^{(i\Delta k - \Delta\alpha)lt_2} \\
& \times \int_0^{t_1} dt_3 e^{(i\Delta k - \alpha_s/2)lt_3} \int_0^{t_3} dt_4 e^{(-i\Delta k - \Delta\alpha)lt_4} \\
& \times \frac{e^{-(p^2 + q^2)a_{42} + 2pb_{42} + c_{42}}}{d_1(t_4)d_8(t, t_1, t_2, t_3, t_4)}
\end{aligned} \tag{73}
$$

where

$$a_{42}(t, t_1, t_2, t_3, t_4) \equiv \frac{d_9(t_1, t_2, t_3, t_4)}{d_{10}(t, t_1, t_2, t_3, t_4)}, \tag{74}$$

$$b_{42}(t, t_1, t_2, t_3, t_4) \equiv W_1 \frac{d_1(t_1)^* g_5(t_1, t_2, t_3, t_4)}{d_{10}(t, t_1, t_2, t_3, t_4)}, \tag{75}$$

and

$$c_{42}(t, t_1, t_2, t_3, t_4) \equiv W_1^2 g_6(t_1, t_2, t_3, t_4) \tag{76}$$

with

$$g_5(t_1, t_2, t_3, t_4) \equiv 2(t_1 - t_2)d_4(t_1, t_3, t_4)^* + 2(t_3 - t_4)d_{21}(t_1, t_3)^* d_2(t_1, t_2) \tag{77}$$

$$
\begin{aligned}
g_6(t, t_1, t_2, t_3, t_4) \equiv\ & \frac{2i\xi_p(t - t_1)g_5(t_1, t_2, t_3, t_4)^2}{d_2(t_1, t_2)d_4(t_1, t_3, t_4)^* d_{10}(t, t_1, t_2, t_3, t_4)} \\
& - \frac{2(t_1 - t_2)^2}{d_2(t_1, t_2)} - \frac{2(t_3 - t_4)^2 d_{21}(t_1, t_3)^*}{d_4(t_1, t_3, t_4)^*}.
\end{aligned} \tag{78}
$$

The functions d_9 and d_{10} are given by

$$d_9(t_1, t_2, t_3, t_4) \equiv 2d_4(t_1, t_3, t_4)^* + d_2(t_1, t_2)d_3(t_3, t_4)^* \tag{79}$$

and

$$d_{10}(t, t_1, t_2, t_3, t_4) \equiv d_2(t_1, t_2)d_4(t_1, t_3, t_4)^* + 2i\xi_p(t - t_1)d_9(t_1, t_2, t_3, t_4) \tag{80}$$

4.5. The fifth order SHG field term

Using Eqns 42, 58, 59 and 19 in 26 and 18, the fifth order pump field term $d^5 E_s^{(5)}$ is obtained as a sum of three terms $d^5 E_{s1}^{(5)}$, $d^5 E_{s2}^{(5)}$ and $d^5 E_{s2}^{(5)}$:

$$d^5 E_{s1}^{(5)}(\mathbf{r}) = \left(\frac{n_s}{n_p}\right)^2 \kappa \mid \kappa \mid^4 E_{p0} f_{s1}^{(5)}(\mathbf{r}) \tag{81}$$

$$d^5 E_{s2}^{(5)}(\mathbf{r}) = 4\left(\frac{n_s}{n_p}\right)^2 \kappa \mid \kappa \mid^4 E_{p0} f_{s2}^{(5)}(\mathbf{r}) \tag{82}$$

and

$$d^5 E_{s3}^{(5)}(\mathbf{r}) = 2\left(\frac{n_s}{n_p}\right)^2 \kappa \mid \kappa \mid^4 E_{p0} f_{s3}^{(5)}(\mathbf{r}) \tag{83}$$

where the expressions for the terms $f_{s1}^{(5)}$, $f_{s2}^{(5)}$ and $f_{s3}^{(5)}$ are given below:

7

4.5.1. $f_{s1}^{(5)}$

$$f_{s1}^{(5)}(\mathbf{r}) \equiv e^{(ik_s - \alpha_s/2)lt} \int_0^t dt_1 e^{(i\Delta k - \Delta\alpha)lt_1}$$

$$\times \int_0^{t_1} dt_2 e^{(-i\Delta k - \alpha_s/2)lt_2} \int_0^{t_2} dt_3 e^{(i\Delta k - \Delta\alpha)lt_3}$$

$$\times \int_0^{t_1} dt_4 e^{(-i\Delta k - \alpha_s/2)lt_4} \int_0^{t_4} dt_5 e^{(i\Delta k - \Delta\alpha)lt_5}$$

$$\times \frac{e^{-(p_a{}^2 + q^2)a_{51} + 2p_a b_{51} + c_{51}}}{d_1(t_3)d_1(t_5)d_{12}(t, t_1, t_2, t_3, t_4, t_5)} \tag{84}$$

where

$$a_{51}(t, t_1, t_2, t_3, t_4) \equiv \frac{d_{11}(t_1, t_2, t_3, t_4, t_5)}{d_{12}(t, t_1, t_2, t_3, t_4, t_5)}, \tag{85}$$

$$b_{51}(t, t_1, t_2, t_3, t_4) \equiv W_1 \frac{g_7(t_1, t_2, t_3, t_4, t_5)}{d_{12}(t, t_1, t_2, t_3, t_4, t_5)}, \tag{86}$$

and

$$c_{51}(t, t_1, t_2, t_3, t_4) \equiv W_1{}^2 \{ \frac{g_8(t_1, t_2, t_3, t_4, t_5)^2 d_{12a}(t, t_1, t_2, t_3, t_4, t_5)}{d_{12}(t, t_1, t_2, t_3, t_4, t_5)_a}$$

$$-g_9(t_1, t_2, t_3, t_4, t_5)\} \tag{87}$$

with

$$g_7(t_1, t_2, t_3, t_4, t_5) \equiv 2(t_2 - t_3)d_1(t_2)^* d_4(t_1, t_4, t_5)$$
$$+ 2(t_4 - t_5)d_1(t_4)^* d_4(t_1, t_2, t_3) \tag{88}$$

$$g_8(t_1, t_2, t_3, t_4, t_5) \equiv 2(t_2 - t_3)\frac{d_1(t_2)^*}{d_4(t_1, t_2, t_3)} + 2(t_4 - t_5)\frac{d_1(t_4)^*}{d_4(t_1, t_4, t_5)} \tag{89}$$

$$g_9(t_1, t_2, t_3, t_4, t_5) \equiv 2(t_2 - t_3)^2 \frac{d_{21}(t_1, t_2)}{d_4(t_1, t_2, t_3)} + 2(t_4 - t_5)^2 \frac{d_{21}(t_1, t_4)}{d_4(t_1, t_4, t_5)}. \tag{90}$$

The functions d_{11} and d_{12} are given by

$$d_{11}(t_1, t_2, t_3, t_4) \equiv d_3(t_2, t_3)d_4(t_1, t_4, t_5) + d_3(t_4, t_5)d_4(t_1, t_2, t_3) \tag{91}$$

and

$$d_{12}(t, t_1, t_2, t_3, t_4) \equiv d_4(t_1, t_2, t_3)d_4(t_1, t_4, t_5)$$
$$+ ik\xi_p(t - t_1)d_{11}(t_1, t_2, t_3, t_4, t_5) \tag{92}$$

4.5.2. $f_{s2}^{(5)}$

$$f_{s2}^{(5)}(\mathbf{r}) \equiv e^{(ik_s - \alpha_s/2)lt} \int_0^t dt_1 e^{(i\Delta k - \Delta\alpha)lt_1}$$

$$\times \int_0^{t_1} dt_2 e^{(-i\Delta k - \alpha_s/2)lt_2} \int_0^{t_2} dt_3 e^{(i\Delta k - \Delta\alpha)lt_3}$$

$$\times \int_0^{t_3} dt_4 e^{(-i\Delta k - \alpha_s/2)lt_4} \int_0^{t_4} dt_5 e^{(i\Delta k - \Delta\alpha)lt_5}$$

$$\times \frac{e^{-(p_a{}^2 + q^2)a_{52} + 2p_a b_{52} + c_{52}}}{d_1(t_3)d_1(t_5)d_{12}(t, t_1, t_2, t_3, t_4, t_5)} \tag{93}$$

where

$$a_{52}(t, t_1, t_2, t_3, t_4) \equiv \frac{d_{13}(t_1, t_2, t_3, t_4, t_5)}{d_{14}(t, t_1, t_2, t_3, t_4, t_5)}, \tag{94}$$

$$b_{52}(t, t_1, t_2, t_3, t_4) \equiv W_1 \frac{d_1(t_1)d_1(t_2)^* g_2(t_2, t_3, t_4, t_5)}{d_{14}(t, t_1, t_2, t_3, t_4, t_5)}, \tag{95}$$

and

$$
\begin{aligned}
c_{52}(t, t_1, t_2, t_3, t_4) & \equiv W_1{}^2 \{ g_3(t_2, t_3, t_4, t_5) + g_4(t_1, t_2, t_3, t_4, t_5) \\
& + \frac{ik\xi_p(t - t_1)d_1(t_1)d_1(t_2)^{*2} h_1(t_2, t_3, t_4, t_5)^2}{d_8(t_1, t_2, t_3, t_4, t_5)d_{14}(t, t_1, t_2, t_3, t_4, t_5)} \}
\end{aligned}
\tag{96}
$$

The functions d_{13} and d_{14} are given by

$$d_{13}(t_1, t_2, t_3, t_4, t_5) \equiv d_1(t_1)d_7(t_2, t_3, t_4, t_5) + d_8(t_1, t_2, t_3, t_4, t_5) \tag{97}$$

and

$$
\begin{aligned}
d_{14}(t, t_1, t_2, t_3, t_4, t_5) & \equiv d_1(t_1)d_8(t_1, t_2, t_3, t_4, t_5) \\
& + ik\xi_p(t - t_1)d_{13}(t_1, t_2, t_3, t_4, t_5)
\end{aligned}
\tag{98}
$$

4.5.3. $f_{s3}^{(5)}$

$$
\begin{aligned}
f_{s3}^{(5)}(\mathbf{r}) & \equiv e^{(ik_s - \alpha_s/2)lt} \int_0^t dt_1 e^{(i\Delta k - \Delta\alpha)lt_1} \\
& \times \int_0^{t_1} dt_2 e^{(-i\Delta k - \alpha_s/2)lt_2} \int_0^{t_2} dt_3 e^{(i\Delta k - \Delta\alpha)lt_3} \\
& \times \int_0^{t_2} dt_4 e^{(i\Delta k - \alpha_s/2)lt_4} \int_0^{t_4} dt_5 e^{(-i\Delta k - \Delta\alpha)lt_5} \\
& \times \frac{e^{-(p_a{}^2 + q^2)a_{53} + 2p_a b_{53} + c_{53}}}{d_1(t_3)d_1(t_5)d_{12}(t, t_1, t_2, t_3, t_4, t_5)}
\end{aligned}
\tag{99}
$$

where

$$a_{53}(t, t_1, t_2, t_3, t_4) \equiv \frac{d_{15}(t_1, t_2, t_3, t_4, t_5)}{d_{16}(t, t_1, t_2, t_3, t_4, t_5)}, \tag{100}$$

$$b_{53}(t, t_1, t_2, t_3, t_4) \equiv W_1 \frac{d_1(t_1)g_5(t_2, t_3, t_4, t_5)}{d_{16}(t, t_1, t_2, t_3, t_4, t_5)}, \tag{101}$$

and

$$
\begin{aligned}
c_{53}(t, t_1, t_2, t_3, t_4) & \equiv W_1{}^2 \{ g_6(t_1, t_2, t_3, t_4, t_5) \\
& + \frac{ik\xi_p(t - t_1)d_1(t_1)h_3(t_2, t_3, t_4, t_5)^2}{d_{10}(t_1, t_2, t_3, t_4, t_5)d_{16}(t, t_1, t_2, t_3, t_4, t_5)} \}
\end{aligned}
\tag{102}
$$

The functions d_{15} and d_{16} are given by

$$d_{15}(t_1, t_2, t_3, t_4, t_5) \equiv d_1(t_1)d_9(t_2, t_3, t_4, t_5) + d_{10}(t_1, t_2, t_3, t_4, t_5) \tag{103}$$

and

$$
\begin{aligned}
d_{16}(t, t_1, t_2, t_3, t_4, t_5) & \equiv d_1(t_1)d_{10}(t_1, t_2, t_3, t_4, t_5) \\
& + ik\xi_p(t - t_1)d_{15}(t_1, t_2, t_3, t_4, t_5)
\end{aligned}
\tag{104}
$$

5. CONCLUSIONS

Expressions for the pump and second harmonic fields are presented as series of terms in increasing powers of the nonlinear optical coefficient. The effects of beam diffraction, linear absorption and beam walk-off are included. An advantage of the method presented here over the widely used numerical method may lie in the increase in the computation speed, especially in special cases. For example, in the case of a collimated pump beam, i.e., for $\xi_p \ll 1$, the single, double and triple integrals in the expressions for the fields reduce to analytical expressions and the quadruple and quintuple integrals reduce to much simpler integrals which can be rapidly evaluated using standard computers. Also, the method can be extended to the case of pump beams of arbitrary shape.

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